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Determining absolute value of thermal neutron flux density based on monocrystalline silicon in nuclear reactors

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Abstract A new type of neutron detector based on monocrystalline Si is developed to measure the fluence and flux density of thermal and fast neutrons. The principle of this detector is based on the relationship between changes in electrical conductivity and neutron fluence during irradiation. Therefore, the absolute values of thermal neutron fluence and flux density are measured in a facile manner with high reliability. Compared with activation methods, our method not only possesses a similar accuracy, but also demonstrates superior application potential for the investigation of neutron fields in nuclear reactors owing to its suitable half-life.

Keywords Thermal neutron · Neutron detector · Neutron flux density · Cadmium difference methods

1 Introduction

In conducting research pertaining to reactors and accelerators, the neutron flux density and the corresponding neutron fluence must be controlled within the ranges of 10^6–10^9 cm^{-2} s^{-1} and 10^{10}–10^{15} cm^{-2}, respectively. However, conventional methods, including proportional counters, ionization chambers, nuclear photographic emulsions, neutron scintillation detectors, and activation detectors, perform unsatisfactorily in this range because of their complex detection processes and/or high cost. In this study, a facile low-cost detector based on monocrystalline Si was developed for neutron control. Monocrystalline Si serves as a semiconductor electronic device in neutron detectors. In addition, neutron transmutation doping (NTD) was performed on monocrystalline Si to obtain a modified semiconductor [1].

In 1967, Blanc reported a neutron detection method using Si transistors [2], which are the predecessors of semiconductor Si detectors. In 1974, Patrikeev developed a method for detecting the fluence of fast neutrons using Si resistors [3]. In 1976, Kramer–Ageev developed a semiconductor detector based on Si diodes and used it to investigate strong gamma neutron fields [4]. The principle of these detectors is based on changes in the electrophysical parameters of Si electrical components under the effect of neutrons. However, the initial electrophysical parameters of these Si electronics are ambiguous, including for the same batch of semiconductor Si, which severely hinders their wide application. Furthermore, after undergoing fast neutron irradiation, the initial parameters of Si electronics can be changed irreversibly, even if high-temperature annealing is performed.

In 1985, monocrystalline Si semiconductors with a standard initial conductivity/resistivity [5] (the resistivity error did not exceed 2% [6]) were developed based on NTD technology by a team from the Tomsk Research Nuclear Reactor (IRT-T). To control the neutron fields in nuclear reactors, Varlachev's group [7] developed a detector based on monocrystalline Si, which operates well for measuring the fluence of fast neutrons. Furthermore, researchers have demonstrated the thermal neutron detection ability of this detector [8]. In the two studies

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mentioned above, results obtained via theoretical calculations and experiments results were compared, and the findings confirmed the high reliability of detectors based on monocrystalline Si [7, 8].

The principle of this detector is based on the relationship between the electrical conductivity of Si and the fluence of neutrons. In Si, radiation defects are formed under the action of fast neutrons. In a semiconductor, the change in electrical conductivity is proportional to the change in defect concentration. Additionally, the change in defect concentration is proportional to the change in the fluence of fast neutrons. Therefore, the change in electrical conductivity is proportional to the change in the fluence of fast neutrons [9]. To measure the fluence of fast neutrons, a simple method based on the principle of an ohmmeter was developed [7].

After the fluence of fast neutrons is determined, the effect of fast neutrons on Si is removed via technological annealing, and the effect of thermal neutrons emerges. Doping with heteroatoms does not alter the metallurgical properties of the crystal considerably but significantly changes the electrical conductivity of the semiconductor. In n-type monocrystalline Si semiconductors, the change in electrical conductivity (compared with the initial electrical conductivity) has been experimentally confirmed to be proportional to the fluence of thermal neutrons during irradiation, with a proportionality factor (K) of $(223.2 \pm 2.2)\times10^{17}\ \Omega\ \mathrm{cm}^{-1}$ [8]. When doping Si samples with a final resistivity of 0.1 Ω cm, they must be irradiated with a neutron fluence of $2.23\times10^{20}\ \mathrm{cm}^{-2}$ such that monocrystalline Si becomes an amorphous solid. Therefore, the registration of the neutron fluence by a neutron detector based on monocrystalline Si can reach the order of magnitude of $10^{19}\ \mathrm{cm}^{-2}$.

The absolute values of the flux density and fluence of thermal neutrons in research reactors must be determined to solve many application and fundamental problems. Some studies[10–12] have shown that threshold activation methods based on activation detectors are self-sufficient, e.g., manganese, cobalt, copper, and gold, since they do not require calibration by other detection methods. In these methods, the relationship between the induced activities of the detectors and the flux density (or fluence) of neutrons is used to define the absolute value of the thermal neutron dose. Currently, they are generally used as reference methods for research reactors. However, these methods are laborious and expensive as they require the use of special equipment. In addition, irradiated samples cannot always serve as tracking detectors for two primary reasons. First, the change in the activity (of the threshold activation detectors) after irradiation depends only on the neutron fluence at the last irradiation time, instead of the full thermal neutron fluence over the entire irradiation period. This implies that after three to four half-lives, the effect of every thermal neutron on the threshold activation detectors will disappear. Second, the neutron fluence cannot be determined accurately when it changes during irradiation. For example, the cobalt detector has a long half-life of up to 5.28 years, which implies that it must be processed as a radioactive material after irradiation. Therefore, a facile low-cost method for measuring the absolute values of the flux density and fluence of thermal neutrons in an IRT-T research reactor using a monocrystalline Si detector is developed in this study.

2 Theory

Similar to thermal neutron measurement, the absolute value of the thermal neutron flux density is determined by the thermal neutron capture reaction. After Si is irradiated by neutrons, atoms of Si-31, which are formed in the radiative capture reaction, transform into the stable isotope phosphorus-31 via β-decay (half-life of 2.62 h). This transmutation impurity can increase the electrical conductivity of n-type monocrystalline Si semiconductors and decrease it in p-type monocrystalline Si semiconductors. The change in the electrical conductivity of monocrystalline Si after irradiation and the annealing of radiation defects are directly proportional to fluence of thermal neutrons [13]. This fact is applied as a basis in methods for measuring neutron flux in relative units [14].

To measure the absolute value of the fluence of thermal neutrons, Si is irradiated with and without a cadmium cylindrical filter, similar to the cadmium difference process, which is typically performed in the threshold activation method. Salient points regarding the method are as follows:

The concentration of phosphorus-31 (C), which is produced during irradiation without a cadmium cylindrical filter, can be expressed in terms of two components: the thermal neutron (C_t) and epithermal neutron (C_{nt}) components.

$$C = C_t + C_{nt} \tag{1}$$

Early research has proven that the concentration (C) exhibits a linear relationship with the change in electrical conductivity and can be expressed as follows:

$$C = (\sigma - \sigma_0)/e\mu_n, \tag{2}$$

where σ_0 and σ are the electrical conductivities of Si before and after irradiation, respectively; e and μ_n are the charge and mobility of electrons, respectively. More importantly, σ must be measured after annealing at 800 °C, at which the effect of fast neutron-induced radiation defects on the change in electrical conductivity is eliminated. Owing to the large absorption cross-section in the thermal neutron region and its rapid decrease in the epithermal neutron region, cadmium-113 is typically used as a thermal neutron absorber.

However, the absorption cross-section of cadmium is not expressible as a step function. Therefore, in cadmium difference methods, the concept of the boundary absorption energy of cadmium (E_{Cd}), which depends on the thickness and shape of the cadmium cylindrical filter, was introduced[15]. It implies that neutrons with energies below E_{Cd} are completely absorbed by the cadmium cylindrical filter, whereas neutrons with energies above this energy are not absorbed. In this case, the resulting error (1%–4%) is compensated for by the cadmium amendment F_{Cd}. Similar to the activation methods, these physical quantities are used in the method developed in this study to measure the absolute values of the flux density and fluence of thermal neutrons. In this approximation, when Si is irradiated in a cadmium cylindrical filter, the epithermal (C_{nt}) neutron component can be expressed as follows:

$$C_{nt} = \int_{E_{Cd}}^{\infty} \Sigma(E)\Phi(E)\mathrm{d}E = C_{Cd}F_{Cd}, \tag{3}$$

where F_{Cd} is a coefficient that considers the weakening of the epithermal neutron field by the cadmium cylindrical filter. The concentration of phosphorus-31 (C_{Cd}) is determined by measuring the electrical conductivity before neutron irradiation (σ_0) and after neutron irradiation (σ_{Cd}):

$$C_{Cd} = (\sigma_{Cd} - \sigma_0)/e\mu_n. \tag{4}$$

Yaryna and Tarnovsky[16] proposed an empirical formula for calculating the E_{Cd} of a cadmium cylindrical filter in isotropic fields, as follows:

$$E_{Cd} = 0.520 + 0.162\ \ln(\xi\ d_{Cd}), d_{Cd} = 0.5 \div 1.5\,\mathrm{mm}, \tag{5}$$

$$\xi = 1.58 - 0.82\ (h/2r) + 0.38\ (h/2r)^2, h/2r = 0.5 \div 1.3, \tag{6}$$

where h and r are the height and radius of the cadmium cylindrical filter, respectively.

In reactors with desirable moderators (water, graphite, beryllium, etc.), the thermal neutron spectrum can be described approximately by the Maxwell distribution. In this case, when using a detector whose reaction cross-section in the thermal neutron region varies according to 1/v (where v is the neutron velocity), the thermal neutron component (C_t) can be expressed as follows:

$$C_t = \chi_t \int_0^{ECd} \Sigma(E)\Phi(E)\mathrm{d}E = \chi_t g_t \Sigma_t \Phi, \tag{7}$$

where Φ is the thermal neutron fluence during the irradiation; Σ_t is the macroscopic cross-section of the reaction ^{30}Si(n,γ)^{31}Si at a neutron energy corresponding to an effective temperature T_{eff}, which is different from the medium temperature T_0 (T_0 is the thermal neutron temperature that conforms to the Maxwell distribution); χ_t is the thermal neutron self-shielding coefficient (the ratio of the number of neutrons that pass through Si to the number of neutrons that enter Si); and g_t is the Westcott factor, which accounts for the difference in the dependence of the (n,γ) thermal neutron reaction cross-section for Si-30 based on the 1/v law. According to the nuclear structure and decay data of the International Nuclear Library Network (IAEA), the (n,γ) thermal neutron reaction cross-section in the region of thermal neutrons adheres strictly to the 1/v law with a Westcott factor g_t of 1 [16]. Because of the capture reaction and the environmental leakage of neutrons, not all neutrons can conform to the Maxwell distribution in the experiment, i.e., $T_{eff} > T_0$ [17].

In particular, when

$$\Sigma_a(kT_0)/\xi'\Sigma_s < 0.2, \tag{8}$$

the logarithmic mean of the energy loss is expressed as

$$\xi' = 1 + \left[(A-1)^2/2A\right]\ln[(A-1)/(A+1)], \tag{9}$$

Σ_a and Σ_s are the macroscopic absorption and scattering cross-sections of the moderator in the moderator, respectively; k is the Boltzmann constant; A is the mass number of the moderator's atom.

$$T = T_0[1 + 0.73A\Sigma_a(kT_0)/\Sigma_s] \tag{10}$$

In this study, for the beryllium moderator, T_{eff} = 1.0066T_0, i.e., T_{eff} is approximately 2 K higher than T_0. Combining Eqs. (1), (3), and (7) yields

$$\chi_t\Sigma_t\Phi = C - C_{Cd}F_{Cd}. \tag{11}$$

Subsequently, the absolute value of the thermal neutron fluence can be expressed as follows:

$$\Phi = \frac{C}{\chi_t\Sigma_t}\left(1 - \frac{F_{Cd}}{R_{Cd}}\right). \tag{12}$$

Combining the formula above with Eq. (2) yields

$$\Phi = \frac{(\sigma - \sigma_0)}{e\mu_n\chi_t\Sigma_t}\left(1 - \frac{F_{Cd}}{R_{Cd}}\right), \tag{13}$$

where

$$R_{Cd} = C/C_{Cd} = (\sigma - \sigma_0)/(\sigma_{Cd} - \sigma_0{}') \tag{14}$$

is the Cd ratio determined based on the electrical conductivity in the experiment. The corresponding thermal neutron flux density φ can be calculated using the thermal neutron fluence Φ and irradiation time τ. The corresponding relationship is expressed as $\varphi = \Phi / \tau$.

3 Experiment

In this study, washers of monocrystalline dislocation-free Si were obtained via the crucibleless zone melting method and used as neutron detectors.

To obtain a monocrystalline Si semiconductor, monocrystalline Si was placed in a fission chamber (type CIT-4) next to the reactor to receive neutrons. When neutrons pass through Si, an increasing number of phosphorus atoms are produced by the transmutation, and the monocrystalline Si becomes an n-type semiconductor. The NTD method, which is based on the neutron capture reaction, enables an extremely uniform dopant distribution to be achieved. For Si, the neutron transmutation doping process is based on the capture of thermal neutrons by Si-30 nuclei (approximately 3.1% of Si comprises Si-30 isotopes). This is similar to the principle of thermal neutron detection based on monocrystalline Si.

The application possibility of the proposed method was substantiated using a threshold activation method to measure the thermal neutron flux density in a research reactor. Experiments were conducted in the HEC-4 channel of the Tomsk Research Nuclear Reactor (with 6 MW of power). The electrical resistivity was measured using the four-probe method, and the measurement error was less than 3% [18]. The thermal neutron fluence was controlled continuously using a standard fission chamber (type CIT-4), which is used in the neutron transmutation doping technique for Si [19]. In this study, the thermal neutron self-shielding coefficient was calculated using the Monte Carlo method [20], in which neutron trajectories were simulated in natural Si. In the simulation, the variable parameters were the thickness and radius of the Si sample. For each group of parameters, we set 10^7 incident neutrons that will be absorbed by Si or ejected out from it.

The calculation results are listed in Table 1; as shown, the thickness d_t and radius r of Si are consistent with those of monocrystalline Si used in the IRT-T. In addition, the effective optical thicknesses d_0 and thermal neutron self-shielding coefficient χ_t are provided in the table.

In this study, the following processes were performed to determine the cadmium ratios of Si R_{Cd}(Si) and gold R_{Cd}(Au).

First, a cadmium cylindrical filter with a height of 10 mm, diameter of 15 mm, and thickness of 1 mm was designed for absorbing neutrons. Second, two Si samples were irradiated in a Tomsk Research Nuclear Reactor. One was irradiated in a cadmium cylindrical filter, and the other was irradiated without the presence of a cadmium cylindrical filter. Before irradiation, the initial resistance of the Si samples was determined using the four-probe method. The distance between the Si samples was 15 cm, and the Si samples were irradiated at 6 MW of reactor power for 4 h. Third, to demonstrate the effect of thermal neutrons on the Si samples, the samples were annealed at 800 °C for 2 h. The temperature of the water tank in the reactor pool was 42 °C, which corresponds to the ambient temperature T_{eff} = 315 K. For comparison, two gold activation detectors were irradiated at 100 kW of reactor power for 10 min. One of them was located in a cadmium cylindrical filter, whereas the other gold activation detector was not. The gold activation detectors were arranged in the same manner as for the Si samples. A schematic illustration of the experiment is shown in Fig. 1.

Table 1 Calculation results of Monte Carlo program [14]

Parameter	Value								
Radius r (cm)	0.5			0.6			0.7		
Thickness d_t (cm)	0.4	0.5	0.6	0.4	0.5	0.6	0.4	0.5	0.6
Thermal neutron self-shielding coefficient χ_t	0.996	0.995	0.995	0.996	0.995	0.994	0.995	0.995	0.994
Effective optical thickness d_0 (cm)	0.583	0.657	0.717	0.634	0.720	0.793	0.677	0.774	0.859
Radius r (cm)	0,8			0.9			1.0		
Thickness d_t (cm)	0.4	0.5	0.6	0.4	0.5	0.6	0.4	0.5	0.6
Thermal neutron self-shielding coefficient χ_t	0.995	0.994	0.994	0.995	0.994	0.993	0.995	0.994	0.993
Effective optical thickness d_0 (cm)	0.715	0.821	0.917	0.747	0.863	0.967	0.777	0.902	1.013
Radius r (cm)	1.1			1.2			1.3		
Thickness d_t (cm)	0.4	0.5	0.6	0.4	0.5	0.6	0.4	0.5	0.6
Thermal neutron self-shielding coefficient χ_t	0.994	0.994	0.993	0.994	0.993	0,993	0.994	0,993	0,992
Effective optical thickness d_0 (cm)	0.803	0.936	1.054	0.828	0.966	1.092	0.850	0.994	1.126
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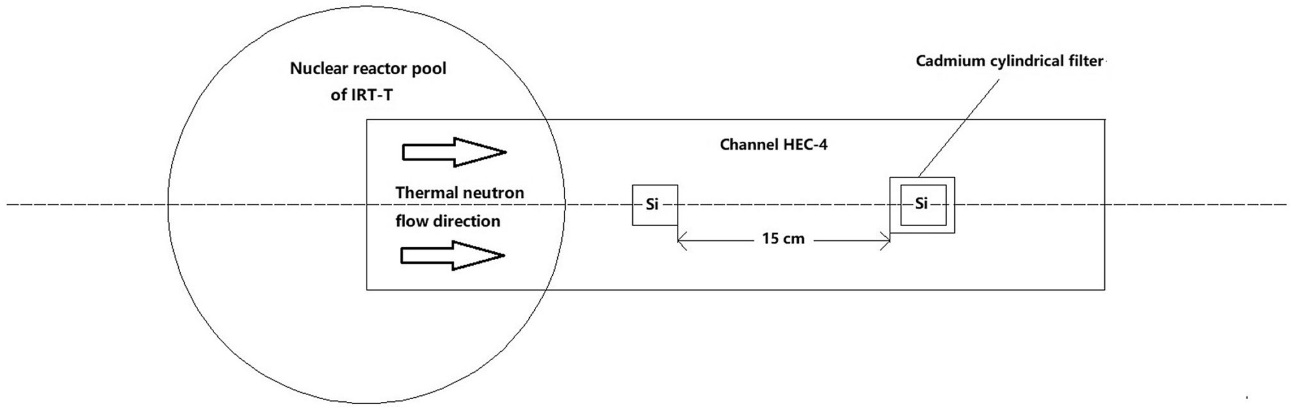

Fig. 1 Schematic diagram of experimental scheme

4 Results and discussion

The resistance of the Si samples was measured using the four-probe method. Based on Eq. (14), the initial electrical conductivity of Si sample A, which is irradiated in the cadmium cylindrical filter, is denoted as σ_0', and that of Si sample B, which is irradiated without the filter, is denoted as σ_0. After irradiation, the final electrical conductivities of the two samples are denoted as σ_{Cd} and σ, separately. The electrical conductivities are listed in Table 2.

The E_{Cd} of the cadmium cylindrical filter can be determined using Eqs. (5) and (6). The boundary absorption energy of this filter was $E_{Cd} = 0.55$ eV.

To calculate the absolute value of the thermal neutron flux density, the following data are required:

(1) Using Eq. 14, the value of the cadmium ratio for Si is $R_{Cd}(\text{Si}) = 16.9$.

(2) The macroscopic section of reaction $^{30}\text{Si}(n,\gamma)^{31}\text{Si}$ can be expressed as follows:

$$\Sigma_t = n\sigma_t, \tag{15}$$

where σ_t is the microscopic section of reaction $^{30}\text{Si}(n,\gamma)^{31}\text{Si}$ and n represents the density of ^{30}Si atoms. In this study, the density of ^{30}Si atoms, n, can be calculated as follows:

$$n = N_A\rho/A, \tag{16}$$

where N_A is the Avogadro constant, ρ is the density of ^{30}Si, g/cm^3, and A is the atomic mass of ^{30}Si (g/mol). The content of ^{30}Si in monocrystalline Si semiconductors is approximately 3.1%. Thus, Σ_t can be calculated as $1.55\cdot10^{-4}$ cm^{-1} (based on a microscopic section σ_t of 0.107×10^{-24} cm^2 listed in the nuclear structure and decay data of the IAEA[16]).

(3) F_{Cd} is typically set to 1.01–1.04[19]. Therefore, F_{Cd} was set to 1.02 in this study (of which the error was 2%).

(4) Based on the calculation results shown in Table 1, the thermal neutron self-shielding coefficient χ_t was set to 0.994.

(5) Before irradiation, the concentration of charge carriers in Si was approximately 10^{14} cm^{-3}. According to Sze, the corresponding electron mobility at this concentration is $\mu_n = 1500$ cm^2 V^{-1} s^{-1} at a temperature of 300 K [21]. In addition, electron mobilities μ_n and $T^{1.5}$ are linearly dependent of each other. Therefore, at the temperature of $T_{eff} = 315$ K, the corresponding electron mobility was $\mu_n = 1613.89$ cm^2 V^{-1} s^{-1}.

(6) The irradiation time for Si samples is $\tau = 14{,}400$ s.

Table 2 Electrical conductivities of silicon samples

Sample	Electrical conductivity (Ω^{-1} cm^{-1})	
	Before irradiation	After irradiation
A	1.17×10^{-3}	1.68×10^{-3}
B	1.30×10^{-3}	1.01×10^{-2}

In summary, the absolute values of the thermal neutron fluence and thermal neutron flux density can be calculated, i.e., $\Phi = 2.07 \times 10^{17}$ cm^{-2} and $\varphi = 1.44 \times 10^{13}$ cm^{-2} s^{-1}, respectively.

The absolute value of the thermal neutron flux density, which was determined via the corresponding method (the gold activation detectors), was 1.45×10^{13} cm^{-2} s^{-1} (converted to a reactor power of 6 MW). The Cd ratio of the gold activation detector was 4.36 [22].

The experimental results show that the absolute value of the thermal neutron flux density measured using Si neutron detectors is similar to that measured using gold activation detectors. Furthermore, the error in the measurement of the absolute value by this method is comparable to that in the threshold activation methods. Therefore, a neutron detector based on monocrystalline Si can be used to calibrate the absolute values of the thermal neutron flux density in any neutron spectrum in nuclear reactors without requiring other calibration methods (e.g., threshold activation methods).

Furthermore, this method is based on changes in physical information (electrical conductivity); therefore, thermal neutron information can be stored indefinitely (unlike threshold activation methods). Compared with cobalt and gold, Si has a more suitable half-life. Hence, Si neutron detectors can be used as a tracking detectors and their radiation results can be verified at any time.

5 Conclusion

We developed a new cadmium difference method based on monocrystalline Si to determine the absolute values of thermal neutron fluence and flux density in nuclear reactors. Our measured results were similar to those of obtained using the conventional method, thus demonstrating the feasibility of the proposed method. Furthermore, this implies that the principle of neutron detection is no longer limited to the detection of secondary charged particles generated by neutron scattering [23, 24] but is based on changes in electrical properties. In addition, semiconductor Si can be obtained readily via neutron transmutation doping during reactor operation [25]. Thus, a neutron detector based on monocrystalline Si is not only a recyclable detector, but can also be custom developed in reactors (which can further control the costs). The proposed method is useful for investigating and controlling neutron fields, particularly in nuclear reactors.

Author contributions V.A. Varlachev contributed to the study conception and design. Material preparation, data collection and analysis were performed by E.G. Emets, Yu-Chen Mu and E.A. Bondarenko. The manuscript was written by V.A. Varlachev and Yu-Chen Mu. All authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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